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TREATMENT OF GASEOUS EFFLUENTS*

By

W. C. Schmidt

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ABSTRACT

Equipment for the removal of noxious radioactive waste gases from chemical processing facilities is described. The discussion is separated into two parts to describe the equipment necessary for the removal of particulate radioactivity present as an aerosol and the other radioactivity which is present as a true gas. As the discussion develops on the decontamination of the waste gases, actual data and empirical equations for design calculations are presented. Also presented is a short section on the difficulties experienced in operating the equipment as described.

TREATMENT OF GASEOUS EFFLUENTS

I. INTRODUCTION

In the operation of any radioactive chemical processing facility, whether it be laboratory, pilot-plant, or full production scale, every effort must be made to prevent the release of harmful amounts of contaminated air or radioactive waste gases to the environment. Ventilation air which passes through a radioactive processing area can entrain matter which, if not removed, may contaminate the environment. The oxides of nitrogen that are given off during uranium dissolution with nitric acid, while not radioactive themselves, do contain quantities of radioiodine in amounts depending upon the radiation history and the cooling time of the metal prior to dissolution. If permitted to discharge to the environment, radioiodine will concentrate in the vegetation and may constitute a health hazard to both livestock and humans. It is important that this isotope be reduced to harmless levels prior to discharge and to the atmosphere.

This paper presents and discusses some of the technological approaches to minimizing both these hazards and will treat the subject in two parts:

1. Decontamination of Ventilation Air by Use of Fiberglas Filters, and
2. The Removal of Radioiodine by the Use of Silver Reactors.

II. DECONTAMINATION OF VENTILATION AIR BY USE OF FIBERGLAS FILTERS

A. Specifications

The removal efficiency requirements to be met in the filtration of radioactive aerosols are more rigorous than those for the ordinary commercial collector. Furthermore, since the collection of large amounts of contaminated material in one place is inherently dangerous, the stringent requirements of the Atomic Energy Commission safety policies restrict

considerably the possible methods by which the air pollution problem may be solved. The specifications which were established as the design goals for ventilation air filters are summarized below. It is evident that the more common types of commercially available dust collectors, such as cyclones and scrubbers, fail to meet one or more of these specifications.

Air Filter Specifications

1. A collection efficiency of 99.99 per cent for submicron particles present in low concentrations.
2. A minimum initial flow resistance commensurate with the desired filtration efficiency.
3. A minimum of maintenance in the continuous operation of the unit.
4. A life expectancy in terms of years.
5. Containment of the collected radioactive materials in a manner that will not cause a subsequent disposal problem.

B. Efficiency of Glass Fibers

The efficiencies with which glass fiber beds remove suspended particulate matter were measured under field conditions by employing as a testing medium the radioactive aerosol discharged from a process cell ventilation system. Particle size determinations, made with modified cascade impactor, indicated that the radioactive particulate matter in the process cell air was predominantly in the submicron range (experimental values of the geometric mean particle varied from 0.2 to 0.7 micron). No attempt was made to determine the size distribution of the uncontaminated particles since the efficiency tests were to be based upon radioactivity measurements. Other studies revealed that the dust loading of the air entering the process cells was 0.01 to 0.02 grains per thousand cubic feet, and that the particle concentration of the effluent air was in the order of 0.2 to 0.4 grains per thousand cubic feet. These figures indicated the preponderance of both the contaminated and the uncontaminated particles of the gas stream originated in the process cell area. It should be noted that the

results which were obtained experimentally are directly applicable only to systems wherein the size distribution and concentrations of the suspended particulate matter are comparable to those of the testing media. In cases where a significant difference in particle size exists, the collection efficiency of a given glass fiber may be appreciably different.

It can be shown in mathematical terms that:

$$DF = CL^aP^bV^c \quad (1)$$

where: DF = decontamination factor, dimensionless
L = filter bed depth, inches
P = packing density, lb/ft^3
V = superficial gas velocity, ft/min
C = a constant, the dimensions of which are such that the decontamination factor is dimensionless
a, b, c = emperically determined exponents.

To determine the exponents for each type of glass fiber, the variance in the collection efficiency was measured in a series of test runs in which only one parameter was varied at a time. Thus, to obtain the velocity exponent, the same bed depth and packing density of a test unit were retained throughout a number of runs, so that the following correlation of efficiency and superficial velocity was in effect:

$$DF = C'V^c \quad (2)$$

and

$$\log DF = c \log V + \log C' \quad (3)$$

By plotting the data on a logarithmic graph of decontamination factor vs. velocity and measuring the slope of the line, the velocity exponent was obtained. This procedure is illustrated in Figure 1, wherein the velocity exponent of Owens Corning "AA" Fiberglas, a fiber having a mean diameter of 1.3 microns, was determined. A similar procedure was employed to determine the exponents for the other parameters as shown in Table I.

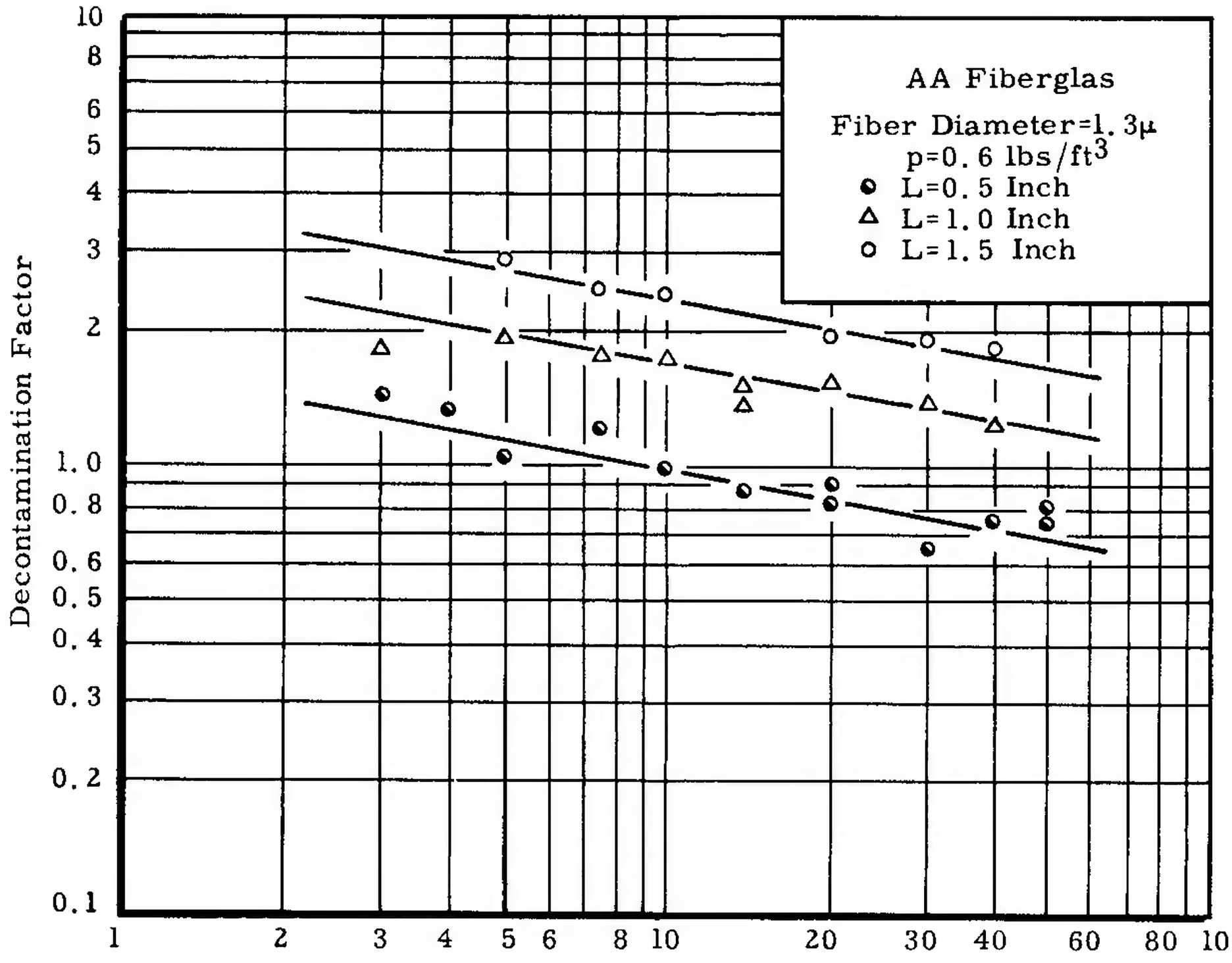

FIGURE 1

Effect of Superficial Velocity on Decontamination Factor

TABLE I

EXPERIMENTAL VALUES OF EFFICIENCY PARAMETERS

Efficiency Parameters for Glass Fibers: $DF = CL^{a}P^{b}V^{c}$

Fiber Code	Fiber Diameter, Microns	C	a	b	c
AA	1.3	4.6	0.8	1.0	-0.2
B	2.5	---	---	---	-0.25
55	15	0.085	0.9	1.1	-0.4
115K	30	0.054	0.9	0.9	-0.4
450	115	---	---	---	-0.5

Throughout the velocity range of 0-75 feet per minute, the collection efficiency of any one type of fiber decreases as the superficial velocity increases, indicating that the predominant mechanisms of particle removal are flow line interception and diffusion of the particles to the fibers with their subsequent adherence. A consistent lowering of the velocity exponent with decreasing fiber diameter is obtained. Since the mechanisms of particle diffusion and particle impingement are influenced in opposite directions by an increase in velocity, and since the effect of particle impingement is greater for smaller diameter fibers, the overall effect should be a flattening of the low velocity portion of the decontamination factor vs. velocity curves and a consequent lowering of the velocity exponent.

Although the data are not correlated on the basis of fiber diameter, it is observed that the efficiency is generally higher for filters having smaller diameter fibers.

C. Resistance of Glass Fibers

The resistance offered by fibrous glass beds to the air streams passing through them was measured in a series of permeability tests in which atmospheric air was employed rather than the contaminated process

cell air. The pressure drop for each type of glass fiber is correlated with the operating variables according to the following relation:

$$\Delta P = KL^x P^y V^z \quad (4)$$

where: ΔP = pressure drop, inches of water
L = bed depth, inches
V = superficial air velocity, ft/min
P = packing density, lb/ft^3
K = a constant
x, y, z = empirical exponents.

The exponents x, y, and z were determined in a series of pressure drops vs velocity measurements for filters which had a different bed depth and packing density. Cross plotting the data results in a general expression for the pressure drop of each type of glass fiber. The data for the fibers which were selected for detailed study are presented in Table II.

TABLE II

EXPERIMENTAL VALUES OF RESISTANCE PARAMETERS

Flow Resistance Parameters for Glass Fibers: $\Delta P = KL^x P^y V^z$

Fiber Code	Fiber Diameter, Microns	K	x	y	z
AA	1.3	0.082	1.0	1.5	1.0
B	2.5	---	---	---	1.0
55	15	0.00043	1.0	1.6	1.0
115K	30	0.00020	1.0	1.5	1.0

The bed depth exponents, as would be anticipated, are one. The velocity exponents are each one, demonstrating that laminar flow exists throughout the flow range (5 to 100 ft/min) which was studied. The value for the packing density exponent is 1.5.

D. Life Expectancy

The proper design of a highly-efficient, fibrous glass filter involves not only an adequate knowledge of the characteristics previously outlined, but also sufficient information regarding the on-stream properties to insure a maximum life expectancy. When a particle is filtered from an air stream, it is assimilated within the bed and causes an incremental change in the operating characteristics of the filter by assisting in the removal of subsequent particles which, in turn, increases the resistance to air flow. The low concentrations of radioactive and inactive particles in the process cell air previously described provided a convenient means of determining the filtration characteristics of fibrous beds under start-up conditions where filtration is due to the fibers alone and the effect of the filtered particles are immeasurably small. The use of the process cell air to determine the comparative life expectancies of glass fiber units was impractical. Therefor, a methylene blue aerosol (0.5 micron size) was employed to provide a high dust loading and thus restrict the test period to a reasonable time interval. It should be noted that, since the exact manner in which the assimilation of collected material continually changes, the operating features of a filter are dependent upon the size distribution, concentration, and structure of the particles. The quantitative results of the methylene blue tests cannot be applied directly to field conditions involving particles of a different nature. The qualitative results, however, are extremely useful in the design of a filter having a maximum life expectancy.

The concept of mechanical filtration as a dynamic process which culminates in the "plugging" of the filter bed was demonstrated by measuring the average collection efficiency and flow resistance during the passage of an incremental amount of methylene blue aerosol to a test filter. In Figure 2 the cumulative grains of dust passing to the unit per square foot of filter area are plotted against efficiency and pressure drop values at a superficial velocity of 25 ft/min for two inches of Owens Corning No. 115K

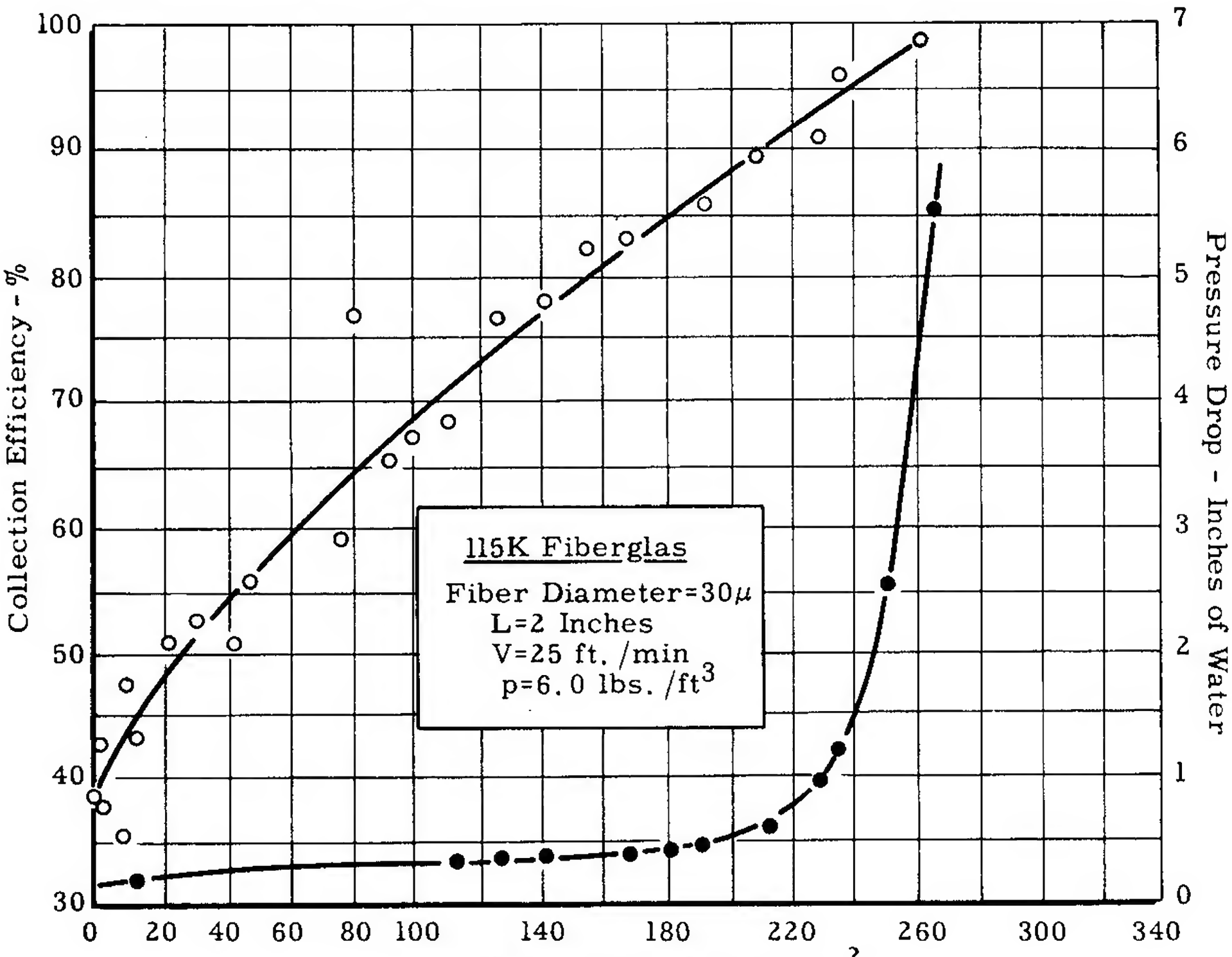

FIGURE 2

Effect of Filter Loading on Pressure Drop and Efficiency

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Fiberglas packed at 6 lb/ft^3 density. The early portion of the pressure drop curve is relatively flat, while the corresponding efficiency value increases rapidly. As the slope of the collection efficiency curve flattens, the pressure drop curve becomes progressively steeper. Thus, the shapes of the two graphs can be interpreted in terms of one another. During the initial rapid rise of collection efficiency, the amount of material assimilated within the bed is not sufficient to bring about an immediate increase in flow resistance. Upon reaching high efficiency values, however, virtually all of the material approaching the unit remains within the bed and, consequently, the pressure drop builds up at a more rapid rate.

To provide sufficient information regarding the on-stream properties of glass fiber beds for development of a highly efficient filter capable of operating for a long period of time, a series of tests were performed wherein the useful filtration lives of various fibrous beds were determined as a function of the quantity of smoke particles passed to the units. The data from the optimum filter are presented in Table III. The data show that if a filter of this composition was incorporated into one filter of laminar type, an expected life of approximately the time required to deposit 3000 grains per square foot of filter area would be obtained.

TABLE III

RESULTS OF LIFE TEST

Type Packing	Density, lb/ft^3	Depth, Inches	Velocity, ft/min	Initial ΔP, Inches of Water	ΔP, Increase Inches of Water	Grains M.B.to unit per sq ft of filter area at ΔP Increase
AA	1.2	0.25	25	0.80	0.13	
55	6.0	8	25	1.58	4.50	125
55	6.0	8	25	1.63	0.20	
55	3.0	5	25	0.33	4.50	300
55	3.0	5	25	0.29	0.08	
55	1.5	14	25	0.27	4.50	3000

E. Stability of Glass Fibers

In addition to the on-stream filtration characteristics of glass fibers, the service life of a filter is influenced by fiber weatherability, i.e., the degree to which the fiber can withstand the effects of the chemical conditions under which it is to be used. The large increase in surface area brought about by the formation of small filaments results in a corresponding increase in its reactivity with acids, alkalies, or water. Weatherability is thus a function of fiber diameter and glass composition. It is advisable to consult the manufacturer concerning the weatherability of a particular type of glass prior to its use in an installation.

F. Final Design of Filter

A convenient method of coordinating and summarizing the material presented in the previous sections is to give an illustrative example of a filter designed to remove suspended particulate matter in a sub-micron range with an efficiency of 99.99 per cent or greater. The filter formulation for the decontamination of the vent gases from a dissolver process vessel is presented in Table IV.

The unit is 2.5 feet by 5.5 feet with an overall height of 4.3 feet. The cross-sectional area, 12.5 square feet, was defined so that under conditions of maximum gas flow the superficial velocity is approximately 20 feet per minute. Weatherability data indicated that the chemical resistivity of the 115K and AA Fiberglas would be satisfactory under the service conditions. The depth and packing densities of the various strata were based upon the relationships discussed above and were selected to provide a maximum service life, a filtration efficiency in the order of 99.99 per cent, and a pressure drop of four inches of water at the rated air velocity.

TABLE IV

FILTER FORMULATION FOR A PROCESS VESSEL

Approximate Superficial Velocity = 20 ft/min.

The gas stream contains varying amounts of sub-micron particles and acid vapors; the latter often reach a volume concentration as high as 50 per cent.

Layer	Packing Type Fiberglas	Bed Density, lb/ft^3	Bed Depth, Inches	Initial Efficiency, Per Cent	Initial Pressure Drop, Inches of Water
Bottom	115K	1.5	12	39	< 0.1
Second	115K	3.0	10	53	0.24
Third	115K	6.0	20	93	1.34
Clean Up	AA	1.2	1	99.9	2.20
Total			43	99.99	3.8

It should be noted that the force necessary to compress large amounts of glass fibers may be large and will influence the structure of containers. The resilience of the material requires that these forces be continually maintained. The necessary compressive force to restrain No. 115K Fiberglas to a given density, for example, is related to the packing density over the range of 1 to 9 lb/ft^3 by the following expression:

$$CF = 2.1 (P - 0.6)^2 \qquad (5)$$

where:

CF = restraining compressive force, lb/ft^2
P = glass fiber packing density, lb/ft^3

This filter formulation was given as an example of a filter for process vessel venting. For a filter to handle large volumes of air such as was described in the ventilation system of a process building, the same packing could be used except the filter area would have to be increased to limit the superficial velocity to 50 feet per minute or less.

G. Operational Difficulties

In actual operations of a fibrous glass filter only minor difficulties have been encountered. The worst of these detriments occur when moisture is allowed to collect in the filter. The AA Fiberglas, which contributes the most to a high removal efficiency, disintegrates somewhat when wet. This disintegration enables the gas to pass through the filter bed without the proper amount of filtration. The other major difficulty encountered is the sealing of the AA Fiberglas to the sidewalls of the container. The 55 and 115K material are somewhat shapeless and very readily assume the size of the enclosing container. The AA Fiberglas, however, is manufactured in mat form and although quite pliable does assume a definite shape. The best method to prevent gas from bypassing the top AA layer along the vessel sidewall is to seal the Fiberglas pad to the sidewall of the container with sodium silicate.

III. REMOVAL OF RADIOIODINE BY USE OF SILVER REACTORS

A. Requirements

The requirements for the removal of the radioiodine are the same as those for the removal of the particulate matter in the ventilation air except that the degree of removal can be relaxed by perhaps a factor of ten. This reduction is possible due to faster rate of decay (half life of 8 days) of radioiodine over the decay rates of the longer-lived fission products that are prevalent in the ventilation air such as zirconium (half life of 65 days) and niobium (half life of 35 days). It is possible to acquire the required decontamination by allowing the irradiated uranium to "cool" until the radioiodine has decayed. However, an economic balance between the cost of "cooling" facilities and the cost of decontamination equipment has to be evaulated for the proper selection of radioiodine removal facilities. An example of the effect of decay time on radioiodine concentration is presented in Table V. These data are theoretically calculated.

TABLE V

IODINE CONCENTRATION AS A FUNCTION OF COOLING TIME

Specific Power KW/lb.	Radioactivity (Curies x 10^{-3}) I^{131}/Ton Natural U Cooling Time, Days						
	0	30	60	75	90	110	150
1	50	3.8	0.28	.076	.02	.0038	.00012
3	150	11.	0.84	.22	.06	.011	.00036
5	250	19.	1.4	.38	.10	.019	.0006
7	350	26.	2.0	.54	.14	.026	.00081
10	500	38.	2.8	.76	.18	.038	.0012
100	5000	380.	28.	7.6	1.8	.38	.012

Note: Radioiodine concentrations indicated for initial concentrations are equilibrium values. That is, the uranium must have been irradiated for at least five and one half half-lives (44 days) for a radioiodine equilibrium to be established in the irradiated uranium.

B. Process

Several different methods of removing the radioiodine from the dissolver off gas were evaluated in the laboratory and pilot plant. Scrubbing with water, trichlorethylene, and ethylene glycol did not provide sufficient decontamination to justify an exhaustive study of these methods. Caustic scrubbing of the gas provided good decontamination; however, as with all types of liquid scrubbing, the subsequent disposal of the contaminated caustic and its related problems were undesirable.

In addition to scrubbing it was found that different treatments of the dissolver solution influenced the amount of radioiodine released to the off-gas. The addition of mercuric ion to the dissolver acid solution to a concentration of 5×10^{-4} M retained 50 - 60 per cent of the iodine in the solution. However, during subsequent processing and especially during a concentration step in an acidic solution, the mercury releases the iodine. On the other hand air sparging of the dissolver solution (in the absence of mercury) during the dissolution of the uranium will remove at least 95 per

cent of the iodine present in the uranium.

Pilot plant experiments indicated that a bed of silver at an elevated temperature would provide the high decontamination factor desired. In addition, such a system would have no subsequent disposal problems of any great magnitude. With these factors in favor of the silver reactor, it is recommended as the best means of removing the radioiodine from the dissolver off-gas.

C. Chemistry and Performance

In the development stages of the silver reactor tests were made on the unit by using an aliquot of the actual radioiodine bearing gas. A portion of the radioiodine entering and leaving the silver reactor was measured by absorbing in a caustic solution which was analyzed for radioiodine. Data from these tests indicated that the retention of radioiodine in the silver bed would be 99.9 to 99.99 per cent. In actual operation, however, the units operate at a retention efficiency of 99 to 99.9 per cent.

The types of silver bearing material which have been used very successfully are: (a) pellets of pure silver approximately 1/16-inch in diameter, (b) a silicon carbide base which contained approximately 3 per cent silver, and (c) unglazed, porous ceramic column packing soaked in a concentrated silver nitrate solutions. Of the three types of packing the silver nitrate coated ceramics were the most economical and for this reason are suggested as the best packing. The unglazed ceramic Berl saddle was selected as the best packing for maximum contact area, minimum pressure drop, and adequate porosity. The porosity of the ceramic controls the absorption of the silver nitrate solution, and as a general criteria, the more silver that is absorbed, the longer the unit remains efficient. For this reason a ceramic that is unglazed and consequently quite porous is desirable.

The largest known contributors to lower efficiencies are low gas temperature and free chlorine in the gas which will replace the iodine

already reacted with the silver. The influence of temperature on iodine retention was further evaluated in the laboratory by passing a ten per cent nitrogen dioxide, ninety per cent air mixture containing iodine over a silver nitrate bed at different temperatures. The results of these tests are given in Table VI. Further investigation in the laboratory also revealed that the amount of iodine passing the silver bed increases approximately as the square of the nitrogen dioxide concentration.

TABLE VI

EFFECT OF TEMPERATURE ON IODINE RETENTION

Air - 10% NO_2
Residence Time in Silver Bed - 1.4 minutes.

Test No.	Temperature, F	Amount of I_2 Passing Bed, micrograms/ ft^3
1	420	.014
2	368	.78
3	320	60.
4	284	580.

Note: 1 microgram I^{131} = 0.125 curies.

In the preliminary stages it was believed that the collection of iodine on the silver reactor could be represented by the equations:

$$6\,AgNO_3 + 3\,I_2 + 3\,H_2O \longrightarrow 5\,AgI + AgIO_3 + 6\,HNO_3$$

which is the reaction in presence of water, or

$$6\,AgNO_3 + 3\,I_2 \longrightarrow 5\,AgI + AgIO_3 + 6\,NO_2 + 3/2\,O_2.$$

Later, experiments in the laboratory have indicated that the following reactions also occur depending upon such factors as nitrogen dioxide concentration, temperature and pressure:

$$2\,AgNO_3 + I_2 \longrightarrow 2\,AgI + 2NO_2 + O_2$$

$$2\,AgNO_3 + I_2 + 2\,O_2 \longrightarrow 2\,AgIO_3 + 2\,NO_2.$$

Thus, it can be seen that an overall chemical equation for the system is very difficult to write because of the changing variables as the uranium-nitric reaction progresses. There is also reported from the laboratory that an isotopic exchange reaction occurs which could partially account for the lowering of actual performance below expected performance:

$$2\,AgI^* + I_2 \rightleftharpoons 2\,AgI + I_2^*,$$

where I* represents radioactive iodine and I is stable iodine.

Several experiments were run in the laboratory by introducing radioiodine over a heated bed of silver iodide, and the amount of radioiodine passing the bed was measured. The indicated efficiency of this reaction was 99.9 per cent. These data indicate that a bed of silver iodide as a final cleanup layer would be beneficial to increase the overall iodine retention efficiency of a silver reactor.

Silver nitrate melts at 413 F which limits the maximum temperature of operation. There is also some evidence of lower melting eutectics of silver nitrate-silver iodide. Melting of this eutectic at operating temperatures might explain the somewhat shorter useful life of the reactors than a stoichiometric calculation would indicate.

D. Description of Equipment

The best piece of equipment to meet the requirements of an iodine removal facility is a tower packed with Berl saddles which have been saturated with silver nitrate. In addition a heat exchanger is required to heat the iodine-bearing gas to the 400 F region before passing it through the silver bearing bed. Temperature measuring instrumentation is required for both the inlet and outlet gas streams. As explained earlier most of the iodine is associated with dissolver off-gas which is highly acidic (as high as 50 per cent nitrogen dioxide) which limits the materials of construction to those which can withstand nitric acid vapors. The tower for containing the saddles could be constructed of type 316 stainless steel and should

be made of sufficient diameter to permit the superficial velocity of the gas to be approximately one foot per second. The recommended height of the packed section is eight feet, but this dimension may be varied somewhat without affecting the overall efficiency.

Both electric and high-pressure steam heaters have been operated quite successfully for heating the radioiodine bearing gas to 400 F. Electric heaters require more maintenance due to burned or shorted elements but are more conducive to maintaining a constant gas temperature. With a sensing element prior to the silver reactor and a proportionating current controller, very close control of the gas temperature can be obtained. On the other hand a well-constructed steam heater requires little maintenance, but steam heaters are less sensitive to changing heat loads. As a consequence the gas temperature varies considerably. One of the most successful heating installations would use both a steam heater for the primary source and an electrical heater of small capacity in series as a booster to keep the gas temperature from oscillating. A schematic diagram of a silver reactor incorporated with a Fiberglas filter is shown in Figure 3.

E. Coating Procedure

In order to apply the silver nitrate coating to the packing, the ceramics are immersed in an 18 to 20 molal silver nitrate solution for one minute, allowed to drip for a short period, and then baked at 220 F for four hours. After the baking, the ceramics are cooled to room temperature and are then ready for insertion into the silver reactor tower.

F. Regeneration Procedure

As mentioned earlier, the expected life of the silver reactor is shorter than a stoichiometric relationship would indicate. Several reasons for this abbreviated life have been postulated among which the most notable are: (a) the formation of silver nitrate-silver iodide eutectics with low melting points allows the silver to drip from the packing; (b) a unimolecular thickness of silver iodide forms on the packing; and (c) large amounts

of other halogens present as impurities in the chemicals used in the dissolver deplete the total silver available for the capture of the radioiodine. To increase this shortened life of the silver reactor, a regeneration technique was developed. The silver reactor is fitted with a spray head at the top of the packing. When gas stream analyses indicate that the reactor is operatting with an impaired efficiency, the unit is allowed to cool to 150 F and the packing is sprayed with 0.04 cubic foot of five molal silver nitrate solution per cubic foot of ceramic packing. After the spraying, the reactor bed is heated to 230 F and allowed to bake for four hours. Again the reactor is cooled and an identical spraying is done. Again the reactor is heated to 230 F and allowed to bake at this temperature for six hours. After the two sprayings and two bake periods, the unit is ready for service. This spraying technique wets approximately fifty per cent of the total packing area without permitting excessive liquid to drain to the bottom of the reactor tower. Reactors have been regenerated with this technique for as many as ten times before the differential pressure becomes excessive for continued use.

G. Operational Difficulties

As a general rule the silver reactors perform very satisfactorily with a minimum of attention or care. This fixed bed adsorption of the radioiodine deletes many problems that would be associated with other methods of removal such as liquid scrubbers. The main difficulty encountered is to assure good control of the bed temperature. If an electrical preheater and sensing thermohm system is used as shown in Figure 3, a sufficient amount of air flow must be maintained at all times to transport the heat from the preheater to the reactor. If the air flow falls below this minimum value, a serious overheating of the preheater will occur. If the reactor is permitted to operate above 413 F, the life of the unit will be seriously affected due to the melting and dripping of the silver nitrate from the column packing. If the reactors are permitted to operate below 375 F, the overall efficiency is lowered. If the temperature is allowed to fall

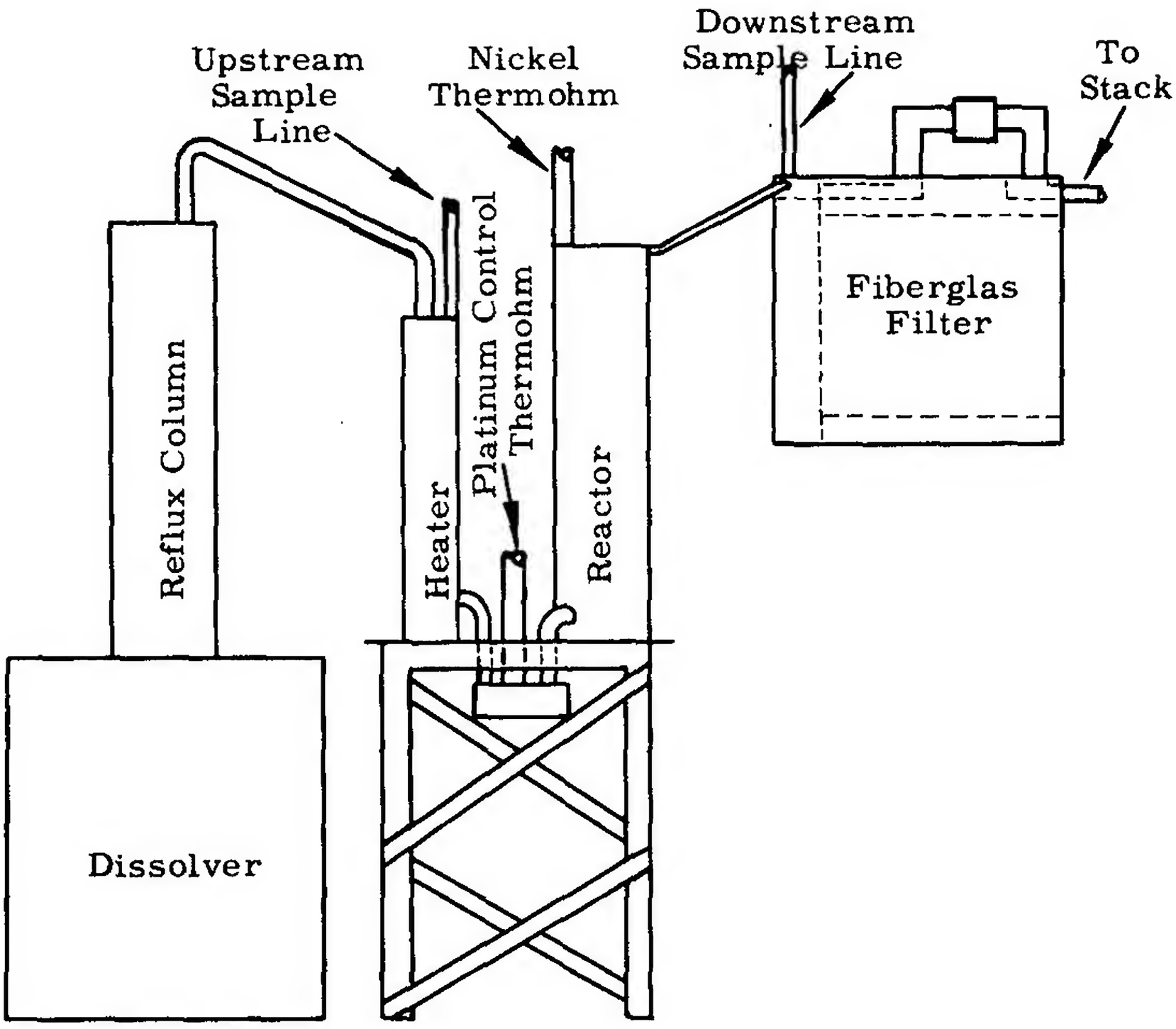

FIGURE 3

Schematic Diagram of Silver Reactor - Fiberglas Unit

below the condensation point of nitric acid vapors (230F), practically no iodine removal occurs. In addition, prolonged operation at this low temperature will deplete the silver bed by the rinsing of the absorbed silver from the packing by the refluxing nitric acid.

IV. REFERENCES

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